REPORT DOCUMENTATION PAGE

AFRL-SR-AR-TR-04-

0132

1. AGENCY USE ONLY *(Leave blank)*	2. REPORT DATE	3. REPOR
	February 25, 2004	Final Technical Report (15 May 99 - 14 May 02)

4. TITLE AND SUBTITLE
Fundamental Interface Structure-Property Relationships for High Temperature Ceramic Composites

5. FUNDING NUMBERS
F49620-99-1-0266
3484/SS
61103D

6. AUTHOR(S)
Elizabeth C. Dickey

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
University of Kentucky
Dept of Materials Science and Engineering
177 Anderson Hall
Lexington, KY 40506-0046

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR/NA
4015 Wilson Blvd., Room 713
Arlington, VA 22230-1954
Program Manager: Dr. Joan Fuller

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION AVAILABILITY STATEMENT
APPROVED FOR PUBLIC RELEASE, DISTRIBUTION IS UNLIMITED

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

Directionally-solidified oxide eutectics such as alumina-YAG and alumina-zirconia show promise as high-temperature structural materials because of their high temperature strength and creep resistance.1-5 Compatibility constraints at the internal interfaces between the two constituent phases can lead to residual stresses upon thermal cycling and elastic interaction stresses under applied loads.6 The magnitudes and distributions of these stresses have important ramifications for the mechanical behavior of the composites. Here we investigate thermal stresses in alumina-YAG and alumina-zirconia directionally solidified eutectics (DSEs). First, the microstructure and crystallography are thoroughly characterized. X-ray diffraction is employed to measure the strain tensors in each phase, which are subsequently converted to stress tensors. Since the experimental measurements provide only average stresses in each phase, anisotropic finite element modeling (FEM) is used to investigate stress distributions in the materials. Comparisons between the experimental measurements and FEM results provide insight into possible stress relief.

20040305 020

14. SUBJECT TERMS

15. NUMBER OF PAGES
8

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
U	U	U	

FINAL REPORT

For AFOSR Award # F49620-99-1-0266

FUNDAMENTAL STRUCTURE-PROPERTY RELATIONSHIPS FOR HIGH-TEMPERATURE CERAMIC COMPOSITES

Elizabeth C. Dickey
Department of Materials Science and Engineering
University of Kentucky

Abstract

Directionally-solidified oxide eutectics such as alumina-YAG and alumina-zirconia show promise as high-temperature structural materials because of their high temperature strength and creep resistance.[1-5] Compatibility constraints at the internal interfaces between the two constituent phases can lead to residual stresses upon thermal cycling and elastic interaction stresses under applied loads.[6] The magnitudes and distributions of these stresses have important ramifications for the mechanical behavior of the composites. Here we investigate thermal stresses in alumina-YAG and alumina-zirconia directionally solidified eutectics (DSEs). First, the microstructure and crystallography are thoroughly characterized. X-ray diffraction is employed to measure the strain tensors in each phase, which are subsequently converted to stress tensors. Since the experimental measurements provide only average stresses in each phase, anisotropic finite element modeling (FEM) is used to investigate stress distributions in the materials. Comparisons between the experimental measurements and FEM results provide insight into possible stress relief.

I. Objectives

This research program aims to elucidate fundamental aspects of interfacial bonding and interfacial compatibility stresses in high-temperature ceramic composites. The principal class of composite materials investigated are directionally solidified ceramic eutectics. To assess the relationship between interface structure and interfacial compatibility stresses in these classes of materials, this research aims to develop analytical techniques to probe the structure and strain state of the material from multiple length scales.

II. Research Results

Microstructure and Crystallography

The YAG - Al_2O_3 DSE fibers were grown by Dr. Ali Sayir of NASA-Glenn via the laser heated floating zone technique. Fibers were drawn with diameters of 80 μm, although the diameter was intentionally increased to 2mm in some sections to increase the sampling volume for x-ray diffraction. The "Chinese script" microstructure of the YAG-Al_2O_3 eutectic, shown in Fig. 1, proved to be uniform across the radius of the fibers as well as among samples taken from different locations along the fiber axis.

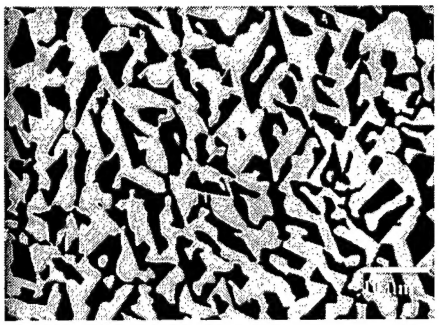

Figure 1. SEM BSE image showing the microstructure of YAG-Al_2O_3 eutectic; YAG is lighter phase.

The crystallographic orientation relationship between YAG and Al_2O_3, shown in Fig. 2, is consistent over the entire cross-section of the sample. The sample normals for Al_2O_3 and YAG are very close to $[\bar{1}2\bar{1}]$ (rhombohedral coordinates) and $[\bar{1}\,\bar{1}1]$, respectively. Perpendicular to the growth axis, the $(2\,\bar{1}1)$ and (011) planes of YAG are coincident with the (111) and $(10\,\bar{1})$ planes Al_2O_3.

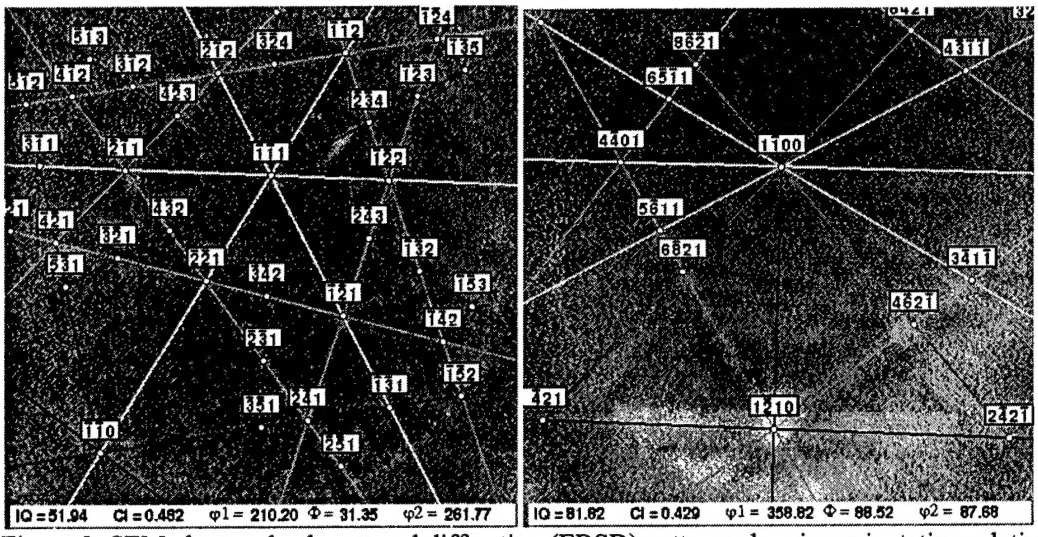

Figure 2. SEM electron backscattered diffraction (EBSD) patterns showing orientation relationship between YAG (left) and Al_2O_3 (right).

This orientation relationship concurs with that found in x-ray diffraction pole figures, as shown in Fig. 3. The pole figures, while showing only one orientation of YAG within the sample, show two twin-related variants of Al_2O_3. The $(10\,\bar{1})$ reflections belonging to each variant are indicated by the black triangles in Fig. 3b.

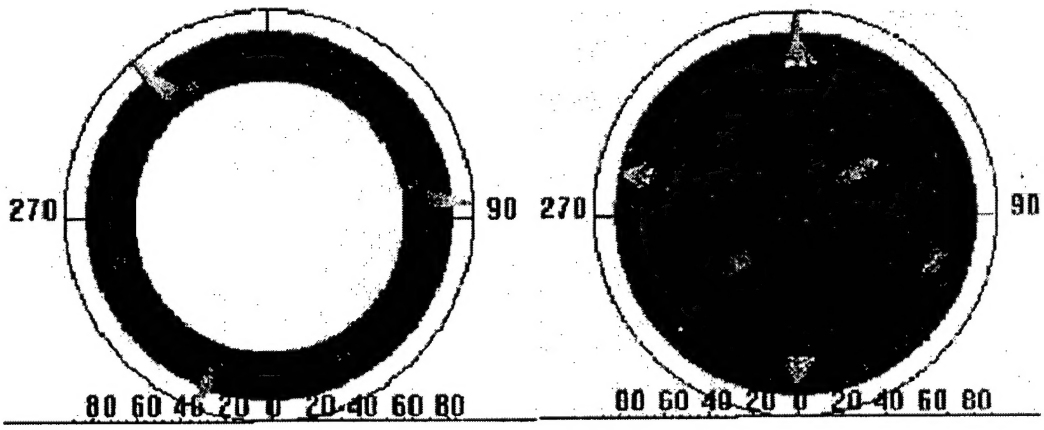

Figure 3. Pole figures done on YAG {111} (left) and Al_2O_3 {10 1} (right) peaks to show orientation relations and variants.

EBSD was used to map the two twin-related variants of Al_2O_3 along the length of a fiber. Fig. 4 shows the spatial variation of the variants. Within what appears to be a contiguous grain, the two twin-related variants are apparent.

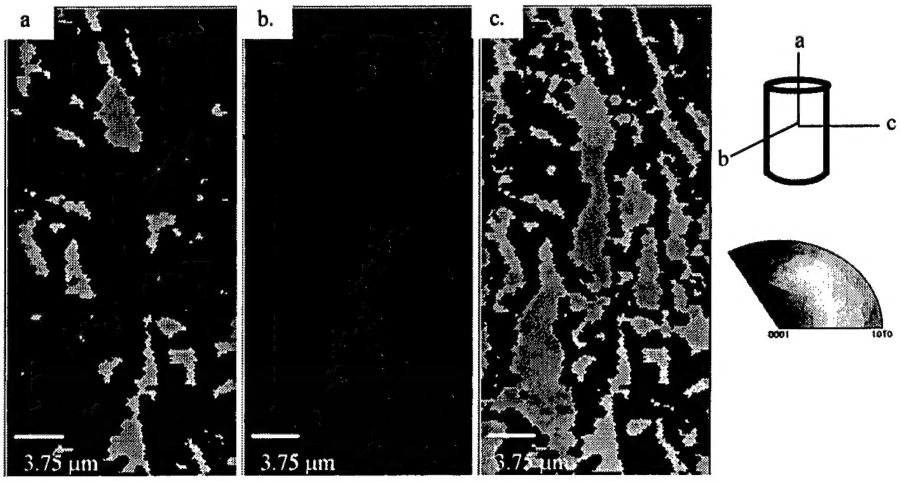

Figure 4 Orientation map showing spatial distribution of the Al_2O_3 variants within a YAG-Al_2O_3 DSE fiber. The sample direction to which the crystallographic directions are referred is indicated in the schematic to the right of the figure.

Fig. 5 shows pole figures from Al_2O_3 and $ZrO_2(Y_2O_3)$ in an Al_2O_3-ZrO_2 DSE. Whereas the Al_2O_3 is nearly single crystalline with [0001] oriented along the growth axis, the ZrO_2 has multiple orientations, although it is highly (220) textured along the growth axis.

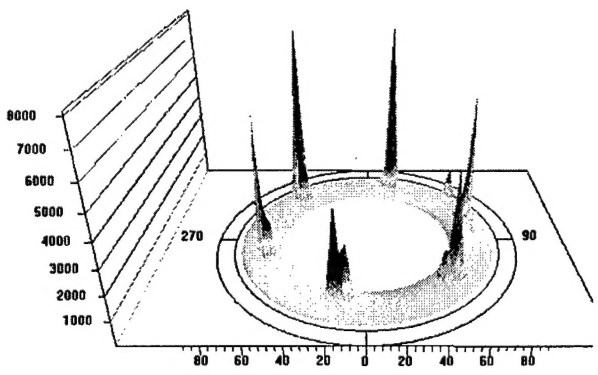

Figure 5a: (1123) pole figure of the Al_2O_3 phase in an Al_2O_3-ZrO_2 DSE. Since the phase is nearly single crystalline, a rotated single crystal stiffness tensor can be used.

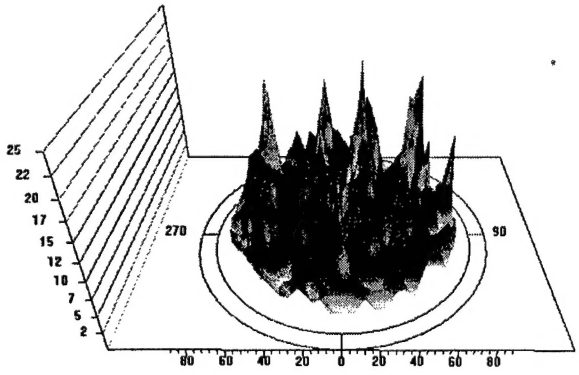

Figure 5b: (311) pole figure of the ZrO_2 phase in an Al_2O_3-ZrO_2 DSE. Since the phase is polycrystalline yet textured, a weighted stiffness tensor must be used.

Measurement of Residual Stresses in Highly Textured Composites by X-ray Diffraction

Because the directionally solidified eutectics are highly textured, standard polycrystalline stress measurements using x-ray diffraction are not possible. It is therefore necessary to make measurements of interplanar spacings along particular sample directions where reflections are present. Once at least six interplanar-spacing measurements have been made and the unstressed lattice parameter measured, it is possible to fit the six components of the strain tensor to the experimental data. Typically, the system is over-determined by making at least twelve measurements and using a fitting routine to determine the strain tensor and error matrix.

The final step in the analysis is to convert the strain tensors to stress tensors with the stiffness tensors. Since the alumina is nearly single crystalline, the single-crystal stiffness tensor rotated to the correct reference frame can be used. The ZrO_2 phase, however, has much weaker texture and it is necessary to weight the stiffness tensor using the orientation distribution function (ODF) as outlined below:

406	105	105	0	0	0
105	406	105	0	0	0
105	105	406	0	0	0
0	0	0	55	0	0
0	0	0	0	55	0
0	0	0	0	0	55

Single crystal stiffness tensor of cubic ZrO_2 (MPa)

+ ODF

336	140	140	-2	-1	2
140	334	141	1	0	-1
140	141	334	0	1	-1
-2	1	0	91	-1	0
-1	0	1	-1	90	-2
2	-1	-1	0	-2	90

Weighted stiffness tensor (MPa)

Applying this procedure to Al_2O_3-ZrO_2(6.6wt% Y_2O_3) DSEs, the following stress tensors were determined where x_3 is normal to the growth axis:

Al_2O_3:

-364	32	56	+/-	9	4	4	(MPa)
	-454	-18			10	5	
		-288				7	

ZrO_2:

2280	-775	8	+/-	20	19	6	(MPa)
	1586	-242			20	6	
		539				11	

The very large stresses, particularly in ZrO_2 which has a lower volume fraction, result from mismatches in thermal expansion/contraction between the two phases and the large temperature differential between the solidification temperature (~2180°C) and room temperature.

In order to measure room temperature residual thermal stress in the alumina-YAG composite, several interplanar spacings of the same crystallographic family of planes in each phase were measured and, in combination with unstressed lattice spacings, strains were calculated and fit to the fundamental x-ray strain equation. The strain tensors were subsequently converted to stress tensors using the single crystal elastic constants. Eighteen measurements were taken of the YAG phase from the family {10 6 4} and eleven measurements were taken of the alumina phase of the families {5 4 3}{2 2 -2}{4 2 0}. The resulting stress tensors and accompanying errors are:

YAG

43	-26	-71			89	36	32
-26	18	-111	MPa	+/-	36	78	40
-71	-111	119			32	40	58

Alumina

296	-70	29			137	34	54
-70	276	65	MPa	+/-	34	136	33
29	65	238			54	33	118

This method of stress measurement was limited by the experimental error, which was on the order of the predicted stresses. It can be seen that most of the stress magnitudes for YAG-Al_2O_3 fall within one standard deviation of error.

Finite Element Modeling of Residual Stresses

To understand stress *distributions* in the materials, we modeled realistic eutectic microstructure by FEM. Microstructural and crystallographic data obtained by scanning electron microscopy (SEM) and electron backscattered diffraction (EBSD) were used to generate models (see Fig. 6). The two phases (YAG and alumina in this case) were completely constrained at the interfaces. Anisotropic thermal and elastic properties were given as inputs to the program. The materials were assumed to be stress free at the eutectic temperature (2100°C) and the temperature was lowered from this point to room temperature.

The results of the FEM calculations are presented in Fig. 7 and show that the YAG is in tension and the Al_2O_3 in compression. This is expected since YAG has a smaller thermal expansion that Al_2O_3. The stresses are largest along the x-axis, corresponding to the c-axis of Al_2O_3. While the average stresses in both phases are low even along the x-direction (~50 MPa in Al_2O_3 and ~-200 MPa in YAG), stress concentrations at sharp corners exceed 600MPa and -550MPa. These areas of stress concentration are the critical areas for mechanical deformation and will certainly be the sites for crack initiation upon loading. Understanding such relationships between microstructure and residual stress is critical for optimizing eutectic microstructures.

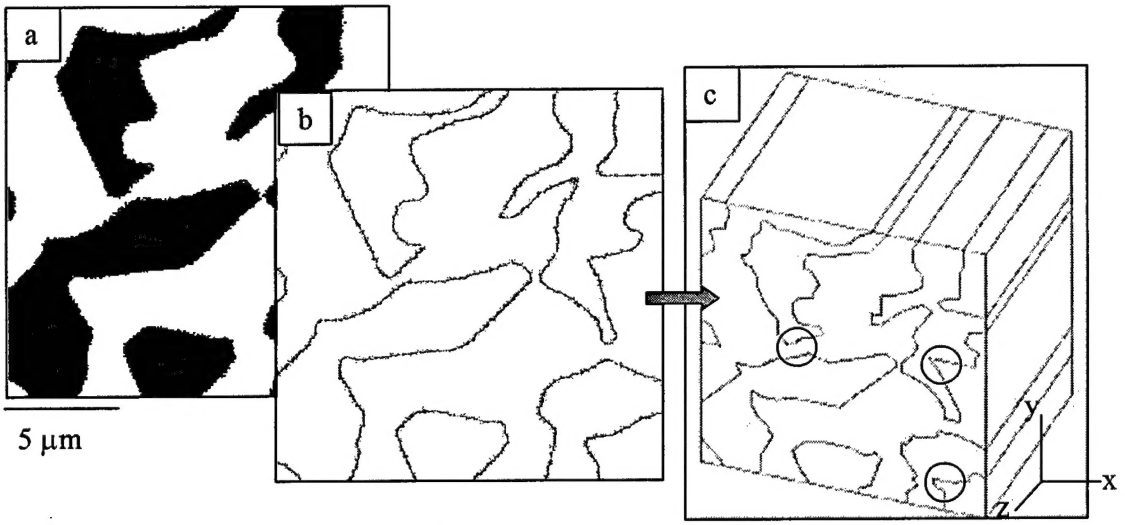

Figure 6: Scanning electron microscope images (a) of the YAG-Al_2O_3 eutectic microstructures are used as the basis for FEM models. The white phase is YAG and the dark Al_2O_3. Boundaries between the phases are identified (b) and the model microstructure extended in the z-direction to make a 3-D model.

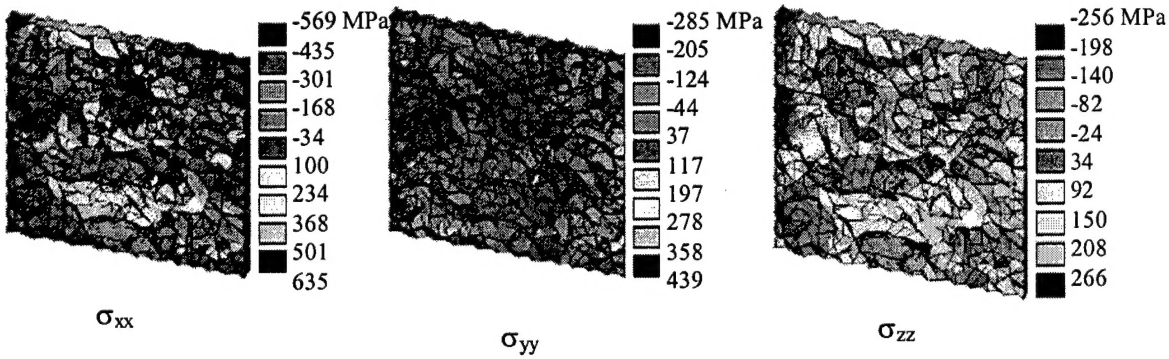

Figure 7: Thermal stresses associated with cooling a YAG-Al_2O_3 DSE from the eutectic temperature (2100°C) to room temperature as calculated by FEM.

References

[1] T. Mah, T.A. Parthasarathy and L.E. Matson, "Processing and mechanical properties of Al_2O_3/Y_3Al_5O12 (YAG) eutectic composite," *Ceramic Engineering and Science Proceedings*, 11, 1617-27 (1990).

[2] T.A. Parthasarathy and T. Mah, "Creep behavior of an Al_2O_3-$Y_3Al_5O_{12}$ eutectic composite," *Ceramic Engineering and Science Proceedings*, 11, 1628-38 (1990).

[3] A. Sayir and L. E. Matson, "Growth and Characterization of Directionally Solidified $Al_2O_3/Y_3Al_5O_{12}$ (YAG) Eutectic Fibers," in HITEMP Review Vol. 1 NASA CP-10082, (1991) 83.1.

[4] A. Sayir, R. M. Dickerson, H. M. Yun, S. Heidger and L. E. Matson, "High Temperature Mechanical Properties of Directionally Solidified $Al_2O_3/Y_3Al_5O_{12}$(YAG) Eutectic Fibers," in HITEMP Review Vol. 1, NASA CP-10146, (1994) 74.1-74.18.

[5] S. C. Farmer, A. Sayir and P. O. Dickerson, "Mechanical and Microstructural Characterization of Directionally-Solidified Alumina-Zirconia Eutectic Fibers," in "Ceramic Matrix Composites-Advanced High-Temperature Structural Materials," eds., R. A Lowden, M. K. Ferber, J. R. Hellmann, K. K. Chawla, and S. G. DiPietro, Mat. Res. Soc. Proc., 365 (1995) 11.

[6] E.C. Dickey, C. S. Frazer, T.R. Watkins and C.R. Hubbard, "Residual Stresses in High Temperature Ceramic Eutectics," *Journal of the European Ceramic Society,* 19 (1999) 2503-2509.

Personnel Supported

This grant supported the research activities of the following people from 1999-2001:

Elizabeth C. Dickey	Assistant Professor, University of Kentucky
Colleen S. Frazer	Graduate Student, University of Kentucky
C. Evan Jones	Student Intern, Dunbar High School, Lexington, KY
Robert Miller	Graduate Student, University of Kentucky

Publications resulting from grant

E.C. Dickey, C. S. Frazer, T.R. Watkins and C.R. Hubbard, "Residual Stresses in High Temperature Ceramic Eutectics," *Journal of the European Ceramic Society,* 19 (1999) 2503-2509.

C.S. Frazer, EC. Dickey, and A. Sayir, "Crystallographic Texture and Orientation Variants in $Al_2O_3/Y_3Al_5O_{12}$ Directionally Solidified Eutectic Crystals," *J. Crystal Growth,* 233 (2001) 187-195.

C.S. Frazer, C.E. Jones, E.C. Dickey, "Interfacial Compatibility Stresses in Alumina-Zirconia and Alumina-YAG Composites," in Advances in Ceramic Matrix Composites VI, *Ceramic Transactions*, Volume 124 (2001) J.P. Singh, Narottam P. Bansal, and Ersan Ustundag, Editors, American Ceramic Society, Westerville, OH, pp.339-350.

Awards Received

Outstanding Materials Engineering Professor, University of Kentucky, 1999 and 2001
Presidential Early Career Award for Scientists and Engineers (PECASE), 1999